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Neutron-deficient polonium isotopes from neon ion bombardments of wolfram

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With 7 figures in the text

Abstract

In a series of experiments, cyclotron-accelerated neon ions have been used to bombard different kinds of wolfram targets. The alpha activities of the reaction products have been studied. Several neutron-deficient polonium isotopes were found, one of which was assigned to ^{196}Po. The half-life was determined to be 1.8 minutes and the alpha particles emitted had an energy of 6.13 MeV. An activity with a half-life of 0.5 minutes and alpha-particle energy of about 6.23 MeV has also been observed. This activity is probably identical with the one earlier assigned to ^{195}Po. Decay data of some neutron-deficient polonium isotopes have been redetermined.

Introduction

At the time when most of the experimental work described in this paper was performed (1956), it was obvious that disagreements occurred among alpha-decay data for neutron-deficient polonium isotopes [1–5] published by different authors. In a plot of alpha-decay energies versus mass numbers for these isotopes the slope of a line through the points was somewhat different if the decay data were taken from, for instance, the study of Karraker *et al.* [4] or if they were taken from the investigation of Rosenblum and Tyrén [5]. In the first-mentioned case, the slope was more in agreement with what could be expected by considering the corresponding data reported for light bismuth and astatine isotopes.

The half-lives of the alpha activities found by the last-mentioned authors were reported as clearly different from each other. Therefore it seemed rather unlikely that some of the alpha peaks in the energy spectrum obtained by them could have been, for instance, due to fine-structure in the decays.

Since April 1955 the 225-cm cyclotron at the Nobel Institute of Physics could produce useful intensities of neon ions (^{20}Ne^{6+} and ^{22}Ne^{6+}) with energies considerably in excess of 100 MeV [6]. In a series of experiments performed by the present authors these ion beams were used to bombard wolfram targets in order to study the neutron-deficient polonium isotopes produced in (Ne, xn) reactions.

As a result of these experiments an alpha activity ascribed to ^{196}Po was found and reported by us in a preliminary communication to the International Conference on

Nuclear Reactions in Amsterdam in 1956 [7]. Later on we also studied light polonium isotopes by bombarding platinum with carbon ions [8, 9]. Tove [10] in 1958 reported four new alpha-radioactive isotopes of polonium, one of which, ^{195}Po, had also been seen by us among the reaction products in the wolfram bombardments. However, a chemical identification of this isotope was not possible in either case because of the short half-life. In the present paper a detailed description of our experiments will be given.

Among other nuclides in this region, Bell and Skarsgaard [11] also studied the EC decay of some light polonium isotopes. Harris [12] has recently given some approximate EC/α-branching ratios of such isotopes produced by heavy ion bombardments in the 60-inch cyclotron in Birmingham, where also earlier investigations in this atomic number region have been made [13].

Bombarding technique

All bombardments were made with internal beams in the 225-cm cyclotron. The heavy ions used were $^{20}Ne^{6+}$ and $^{22}Ne^{6+}$ produced by feeding the ion source with neon gas which in the last-mentioned case was highly enriched in ^{22}Ne. The bombardments with ^{20}Ne ions were usually performed at a radius of 80 cm and those with ^{22}Ne ions at 76 cm. In both cases the nominal energy was about 170 MeV. However, owing to the shape of the continuous energy distribution for the 6-charged ions [6] the majority of the particles had much lower energies. For the penetration of the Coulomb barrier in the actual bombardments neon energies of at least 100 MeV were needed. Foil absorption measurements at a radius of 80 cm indicated that roughly half the total intensity of ^{20}Ne ions having energies above 100 MeV was due to ions with energies in the range 100–120 MeV. However, it should be noted that the energy distribution most likely was somewhat different in different bombardments due to variations in the cyclotron operating conditions.

In the bombardments of natural wolfram, thick targets were used. These consisted of metallic foils, 25 μ thick, clamped to a probe as shown schematically in Fig. 1.

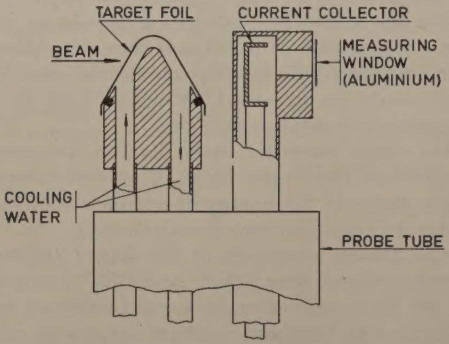

Fig. 1. Simplified representation of the foil probe used in bombardments of thick targets. For measuring the ion current the probe is turned through 180° to bring the measuring window and the insulated current collector into the beam.

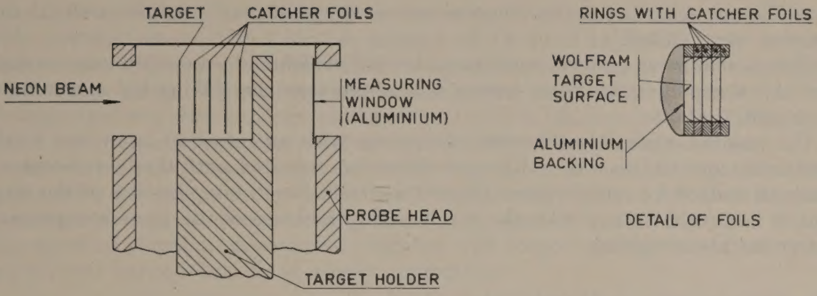

Fig. 2. Schematic drawing of the foil arrangements used for collecting recoiling products. Some components have been omitted for clarity. The probe can be turned through 180° to bring the measuring window into the beam. The probe opening facing the beam is a circular hole 8 mm in diameter.

The arrangement was such that efficient cooling was achieved by a water-flow in direct contact with the back of the target foil.

After the bombardment the irradiated part of the foil (13×19 mm^2) was quickly removed from the probe by cutting. The withdrawal of the probe and removing of the foil was usually finished within about half a minute after the end of the bombardment.

When bombarding isotope-enriched wolfram targets (90 per cent in ^{182}W and ^{184}W respectively) the recoil technique was used. For this purpose the target was prepared as a thin layer of a wolfram compound painted on a backing consisting of a nickel foil, about 1 μ thick. The preparation of the target layer is described below.

The target foil was attached to an aluminium ring (8 mm in inner diameter) which was clamped to a water-cooled target holder inside a water-cooled probe. Catcher foils of aluminium or Tygon mounted onto supporting aluminium rings by zapon were clamped behind the target foil. The aluminium catcher foils were prepared by an evaporation procedure described earlier [14]. The foil arrangement is shown in Fig. 2. The detachment of the target holder could be accomplished in approximately half a minute after the end of bombardment. A detailed description of the probes and the bombarding technique used is published elsewhere [6].

Beam currents were usually measured for ions with energies greater than about 120 MeV. These intensities were in the range 10^{-3}–10^{-2} μA. The irradiations lasted from a few minutes to about two hours.

Chemical procedures

The preparation of the thin wolfram targets

In those bombardments in which the reaction products were collected by the recoil technique very thin wolfram targets had to be used. Several attempts were made to electrodeposit wolfram onto the very thin nickel foil which was to be used as target support. These experiments were unsuccessful. Later work showed that a useful procedure was the following one.

Wolfram oxide was dissolved in a small amount of ammonia heated in a water bath. To one milliliter of this solution several drops of partly polymerized ethylene diamine were added.

The mixture was painted onto the nickel foil, which had previously been mounted (by use of zapon or Araldite) onto a thin aluminium ring fitting the target holder as described above.

The painted surface was heated to dryness with an infra-red lamp and further cautiously ignited in an oven. The procedure was repeated until the layer became as thick as wanted for our purpose (about 2 mg/cm^2). Small lose particles of the target had to be brushed away from the surface. The thickness of the layer was generally determined by weighing.

Chemical separation and identification of polonium

The chemical identification of the radioactive products was made after thick-target bombardments as will be described below. When the recoil collection technique was used most often no chemical separation of the reaction products in the catcher foils was performed. In these cases the activities were identified by their decay properties known from the experiments where chemical procedures had been used.

The polonium isotopes assigned to the mass numbers 196–198 were identified chemically only by an evaporation procedure in which polonium could be separated from bismuth and most other elements. As mentioned above, the activity ascribed to ^{195}Po had not been separated from other reaction products formed in the bombardments.

Chemical separation of polonium from thick wolfram targets.—The thick targets of natural wolfram used in several irradiations were treated chemically after the bombardments.

Several methods were studied in attempts to overcome the chemical difficulties involved by the presence of wolfram and the hydrofluoric acid necessary to dissolve the wolfram target [15]. The procedures used to remove the hydrofluoric acid rapidly after the dissolution of the target caused the wolfram to be precipitated as hydrated oxide, carrying with it some of the polonium.

Two procedures used for rapid separation of polonium from the thick wolfram targets were successful. Thus, volatilization after dissolving the target surface with a few drops of acid or electroplating from solutions containing hydrofluoric acid could be applied.

The volatilization method.—The target foil was heated under an infra-red lamp immediately after the end of the cyclotron bombardment. A few drops of a solution, 9 N in HF and 2 N in HNO_3, were pipetted onto the target surface and spread over the entire bombarded area. During the short time of solvent evaporation, the wolfram surface dissolved, giving a thin salt layer. The dried target was then placed on a grid support and the polonium was evaporated from the heated target onto a platinum plate, which was cooled on the rear side with a small piece of moist tissue.

An evaporation yield of up to about 50 per cent could be obtained if the target was heated to a faint red colour for about 30 seconds. To prepare very thin alpha-radioactive tracer samples, it is recommended that the distillation be repeated.

The electrolytic deposition method.—For the dissolution of the target, 1.0 ml of 11 N HF and 0.2 ml of 14 N HNO_3 were used per millimole of wolfram. In order to keep the metal in solution during the electrolysis, 2 ml of a 1 % tartaric acid solution was added. Finally, the mixture was diluted to 20 ml per millimole of wolfram, and the acidity increased by a further addition of 0.8 ml of 14 N HNO_3. Of course, the amounts and the concentrations of the solutions mentioned here could be changed within certain limits. Experiments showed [15] that a convenient and rapid electrolytic separation was obtained from solutions 0.2–0.5 N in HF, 0.4–0.7 N in HNO_3 and 0.1 % in tartaric acid, for a concentration of 50 millimoles wolfram per litre.

In the electrolytic cell platinum electrodes were used. The cathode was a round disk (diameter 2.5 cm), painted on one side with nail-polish to ensure polonium deposition on one side only. A current density of about 50 mA/cm^2 was used in order to get a high speed of polonium deposition. Working with longer-lived polonium isotopes, a much lower current density is usually preferable.

Counting technique

The alpha activity of the samples was investigated by means of two different types of detectors and counting equipment. In cases where the energy of the alpha particles was of prime interest a combination of a grid ion chamber and a 50-channel pulse-height analyzer was used. These instruments have been described elsewhere [16]. The most short-lived activities could not be measured in that way because it took at least one minute to bring the sample into the ion chamber before counting could be started. In the search for short-lived activities a scintillation detector was therefore used. In this case no energy determination was possible. A schematic drawing of this detector is shown in Fig. 3. The counting rate versus total multiplier voltage (plateau curve) using a ^{210}Po source ($E_\alpha = 5.30$ MeV) is also shown. The load resistor for the collector of the multiplier was 220 kΩ and the sensitivity of the

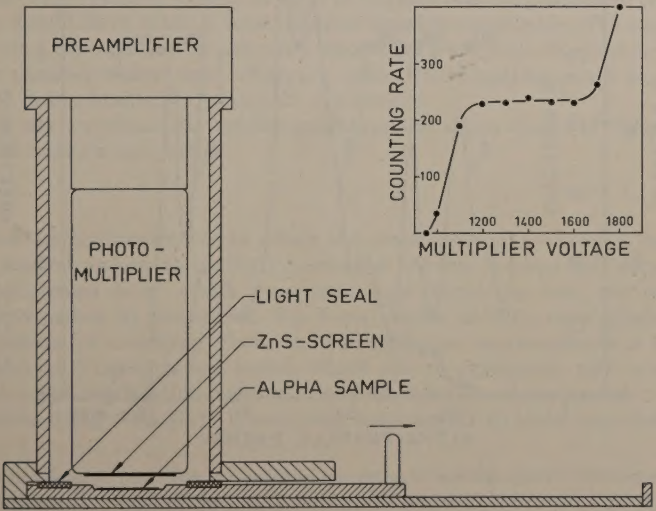

Fig. 3. Schematic drawing of the scintillation detector for alpha particles and operating characteristic for 5.3 MeV alpha-particle energy.

scaler following was 1 volt. The alpha-sensitive layer was prepared directly on the surface of the photomultiplier by sedimenting 5 mg/cm^2 of silver-activated ZnS from an amyl acetate slurry. The efficiency of the arrangement corresponded to a solid angle of about 15 %.

By using the vaporized aluminium catcher foils also as samples, the alpha-measurements could be started very soon after the end of a bombardment. However, the energy spectra obtained by the pulse analysis of the alpha activities in these foils had generally not a very high resolution. The reasons were straggling in the 50–100 μg/cm^2 thick aluminium layer and interference by the large amounts of beta activities from the reaction products collected in the foils.

A more accurate although not so fast method was later found by use of Tygon films as catchers. Quite thin and invisible alpha samples were prepared in this way as has been described by us elsewhere [8, 9].

As standard samples for the energy calibrations ^{210}Po (5.30 MeV), ^{241}Am (5.48 MeV) and ^{242}Cm (6.11 MeV) were used.

Results and discussion

For the half-life determinations and the calculations of the yields of the different polonium isotopes the energy spectra obtained during different time intervals were

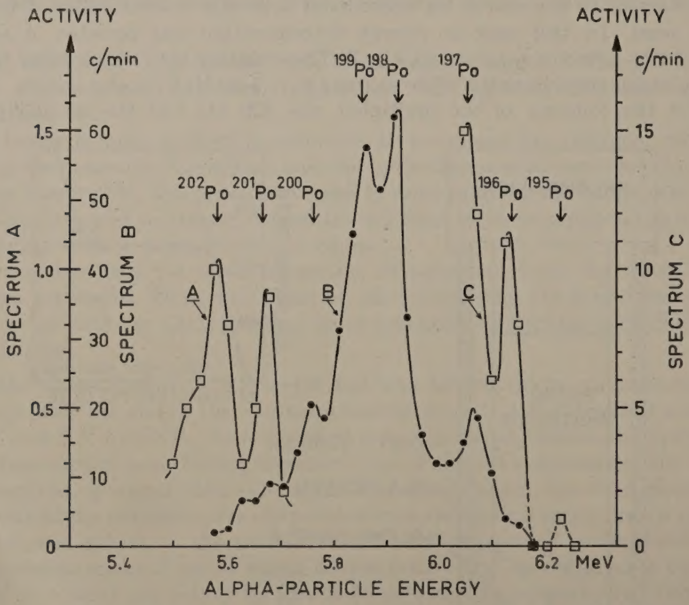

Fig. 4. Alpha-particle energy spectra of polonium isotopes. *A*, spectrum obtained in the interval 50–60 minutes after the start of counting from a thick target bombardment. *B*, spectrum obtained during the first two minutes after the start of counting from the same bombardment as in *A*. *C*, spectrum obtained during the second minute after the start of counting in a recoil experiment; part of the curve is dashed in order to indicate the statistical uncertainty in that region.

plotted from the 50-channel pulse-height analyzer data. In most cases there were large overlaps between the different monoenergetic alpha-particle groups in the spectra as seen in Fig. 4. However, these groups could be resolved by assuming that each peak in the spectrum had the same general shape which was rather close to a Gaussian curve. Some deviation from this type of distribution had to be taken into account on the low energy side of the intensity maximum. The half-life determinations were then made by measuring the area of these resolved groups in the spectra.

In some cases it has been necessary to make corrections for decay during the times of observation. This was made according to the equation

$$A_t = \bar{A}\,\frac{\lambda\tau}{1-\exp(-\lambda\tau)}$$

(cf. e.g. [17]), in which A_t is the true activity value at the time t, i.e. at the beginning of an observation time τ. $\bar{A}$ is the average value of the activity over the time interval τ, and λ is the decay constant. In those cases where $\tau < t_{\frac{1}{2}}$, $\bar{A}$ has been used as a good approximation for $A_{t+\frac{\tau}{2}}$ according to the relation

$$A_{t+\frac{\tau}{2}} = \bar{A}\,\frac{\lambda\tau}{\exp(\lambda\tau/2)-\exp(-\lambda\tau/2)}.$$

Even for $T = t_{\frac{1}{2}}$ the correction which had to be applied to $\bar{A}$ is only -2%. This is a rather small value compared with the statistical errors at the relatively low activities obtained in the experiments in this paper.

In Fig. 4 three alpha-particle energy spectra are shown. All curves are drawn through the experimental points in spite of the fact that the statistical errors in the intensities should have made a more flexible drawing reasonable. The experimental points in a single spectrum were generally too few for good peak energy determinations due to the channel widths used. However, such determinations could be done from the bulk of data obtained in about 35 irradiations.

Some of our experimental results regarding the alpha decay of light polonium isotopes are summarized below.

^{195}Po

In two of the bombardments in which the recoil technique was used, the counting could be started before the activity ascribed to this isotope had decayed down to the background level which was about one count per hour per channel. In the run represented by curve C in Fig. 4 six counts of ^{195}Po were obtained during the first minute of counting. From the ion chamber measurements a half-life of approximately 0.5 minutes was found, which was in agreement with a short-lived component in the scintillation counter measurements. The alpha-particle energy was determined as 6.23 ± 0.04 MeV. These data were similar to those reported by Tove [10].

^{196}Po

Decay data for this isotope were first given by us in the preliminary report to the Amsterdam Conference in 1956 [7]. The data have been slightly changed

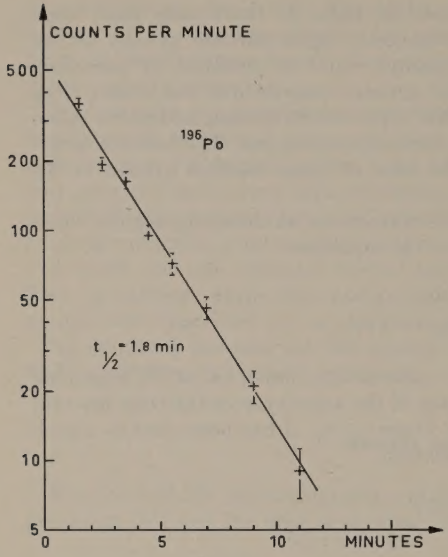

Fig. 5. Decay curve of ^{196}Po from alpha particle pulse-height analysis.

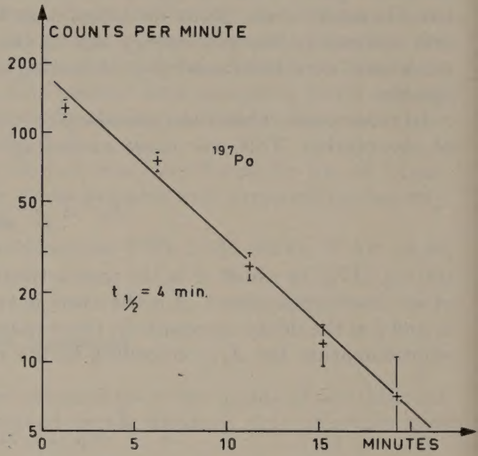

Fig. 6. Decay curve of ^{197}Po from alpha particle pulse-height analysis.

since then by the results of our experiments in the later part of 1956. Thus, the alpha-particle energy is now given as 6.13±0.03 MeV and the half-life as 1.8 minutes. A decay curve is seen in Fig. 5. The value reported by Tove were 6.10 Mev and 110 seconds [10].

^{197}*Po*

The decay data given by Rosenblom and Tyrén [5] were confirmed within our experimental errors. Our alpha-particle energy is 6.05±0.03 MeV and the half-life was close to 4 minutes as is seen from Fig. 6. The energy value is slightly higher than that obtained by us from carbon bombardments of platinum [8, 9].

For the polonium isotopes in the mass number range 198–202 similar results were obtained as those already reported by us [8, 9] from the carbon ion bombardments of platinum. A half-life determination for ^{199}Po is shown in Fig. 7. The decay data for the other isotopes in this range were generally better determined in the last-mentioned experiments than in the present investigation. It should be pointed out that also in this study the half-life found for ^{202}Po is much shorter than has earlier been reported by other authors. A similar result has been obtained by Harris [12].

The experimental conditions influencing the relative yields of the polonium isotopes might have been different in different experiments. For these reasons, amongst others, no reliable quantitative comparisons of the relative yields from different bombardments could be made. However, in some cases expected quantitative relations were obtained. Table 1 gives the relative yields of the polonium alpha activities produced in two bombardments with ^{20}Ne ions using as targets ^{182}W and ^{184}W,

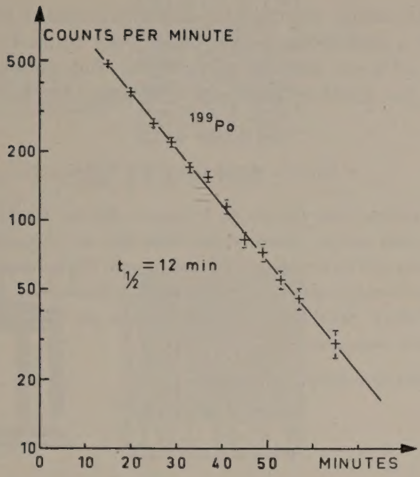

Fig. 7. Decay curve of ^{199}Po from alpha particle pulse-height analysis.

respectively. It can be seen from this table that the difference in the relative yields is at least qualitatively what could be expected if the excitation energies of the compound nuclei formed have been about the same in both cases.

The possible influence of bismuth alpha activities on the alpha-particle energy spectrum of the polonium isotopes has been discussed in detail in two previous papers [8, 9]. From our experiments the possibility cannot be entirely excluded that one of the short-lived polonium isotopes could have decayed by electron-capture to a still more short-lived alpha-emitting bismuth isotope. The first-mentioned nuclide should in such a case mainly contribute to the half-life found, the second one should have the alpha-particle energy measured from the ion chamber runs. However, from

Table 1. Alpha saturation activities of polonium isotopes produced by ^{20}Ne ion bombardments of ^{182}W and ^{184}W respectively.

The activities are given in units of that obtained for ^{196}Po in each of the two bombardments.

Po mass number	Relative activities	
	Bombardment of ^{182}W	Bombardment of ^{184}W
196	1.0	1.0
197	0.7	2.3
198	0.2	0.4
199	<0.1	0.4
200	<0.1	0.1
201	<0.1	<0.1
202	—	<0.1

what is known to date about the modes of disintegrations of light polonium and bismuth isotopes such a possibility seems to be very unlikely.

Finally, it could be of some interest to mention that polonium now seems to be the element of which the greatest number of isotopes are known.

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REFERENCES

1. D. H. Templeton, J. J. Howland, and I. Perlman, Phys. Rev. *72*, 758 (1947).
2. E. L. Kelly and E. Segrè, Phys. Rev. *75*, 999 (1949).
3. D. G. Karraker and D. H. Templeton, Phys. Rev. *81*, 510 (1951).
4. D. G. Karraker, A. Ghiorso, and D. H. Templeton, Phys. Rev. *83*, 390 (1951).
5. S. Rosenblum and H. Tyrén, Compt. Rend. *239*, 1205 (1954).
6. H. Atterling, In course of publication.
7. H. Atterling, W. Forsling, and B. Åström, Proceedings of the International Conference on Nuclear Reactions, Amsterdam 1956; Physica *XXII*, 1193 (1956).
8. H. Atterling, W. Forsling, and B. Åström, Proceedings of the Conference on Reactions between Complex Nuclei, Gatlinburg, Tenn., U.S.A., May 5–7, 1958; ORNL-2606, p. 248.
9. H. Atterling and W. Forsling, Arkiv för fysik *15*, 81 (1959).
10. P. A. Tove, Arkiv för fysik *13*, 549 (1958).
11. R. E. Bell and H. M. Skarsgaard, Can. J. Phys. *34*, 745 (1956).
12. R. G. Harris, Private communication.
13. W. E. Burcham and B. C. Haywood, Proc. Phys. Soc. *A 69*, 862 (1956).
14. R. B. Leachman and H. Atterling, Arkiv för fysik *13*, 101 (1957).
15. G. Oelsner and W. Forsling, Arkiv för kemi *11*, 349 (1957).
16. B. Åström, Arkiv för fysik *12*, 215 (1957).
17. D. Taylor, The Measurements of Radio Isotopes, p. 11. London, 1951.

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